

# Triplet superconductivity in the skutterudite $\text{PrOs}_4\text{Sb}_{12}$

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(Dated: February 2, 2008)

## Abstract

There is mounting evidence for triplet superconductivity in the recently discovered skutterudite compound  $\text{PrOs}_4\text{Sb}_{12}$ . In this work, we propose nodal order parameters for the A- and B-phases of this superconductor which are consistent with angle dependent magnetothermal conductivity measurements and with low-temperature thermal conductivity data in the range  $T \gtrsim 150$  mK. The quasiparticle density of states and the thermal conductivity  $\kappa_{zz}$  are derived within the quasiclassical approximation.

PACS numbers:

## 1. Introduction

The skutterudite compound  $\text{PrOs}_4\text{Sb}_{12}$  is a heavy-fermion superconductor with a transition temperature of 1.8K. [1, 2, 3] Angle dependent magnetothermal conductivity measurements on this material have revealed an interesting multi-phase structure, characterized by energy gap functions  $\Delta(\mathbf{k})$  with point nodes.[4, 5] Previously, several unconventional order parameters, including s+g-wave symmetry, have been proposed to account for this nodal structure. [6] Most of these models that have so far been considered for  $\text{PrOs}_4\text{Sb}_{12}$  are not able to describe self-consistently the observed angle dependence.[7]

Recently, there has been mounting experimental evidence for triplet superconductivity in this compound. First, from  $\mu$ -SR measurements Aoki et al. discovered a remnant magnetization in the B-phase of this compound, indicating triplet pairing.[8] Second, thermal conductivity measurements along the  $[0\ 0\ 1]$  direction at low temperatures ( $T \gtrsim 150$  mK) indicate T-linear and H-linear behavior, consistent with a triplet order parameter.[9] Third, the observed angle dependence of  $\kappa_{zz}$  with  $\vec{H}$  rotated within the x-z plane indicates triplet superconductivity. Finally, recently reported NMR data by Tou et al.[10] for the Knight shift in  $\text{PrOs}_4\text{Sb}_{12}$  also suggest triplet pairing.

The objective of this paper is twofold. First, we propose a new set of nodal order parameters for the A and B phases of  $\text{PrOs}_4\text{Sb}_{12}$ , which are able to describe the thermal conductivity data observed in Refs. [4] and [9]. The proposed model can also describe the isotropic superfluid density in the B-phase reported by Chia et al.[11], albeit with the provision that it has to be assumed that the nodal points in this experiment are aligned parallel to the external magnetic field when the sample is field-cooled. [12, 13] In order to interpret the observed  $\theta$ -dependence of  $\kappa_{zz}$  [9] in terms of the present model, the nodes in the B-phase have to be aligned parallel to  $[0\ 0\ 1]$ .

The second objective of this work is to study and make predictions for the nodal excitations in the vortex state of the proposed model. Simple expressions will be derived for the quasiparticle density of states as well as for the angle dependent magnetothermal conductivity. The thermal conductivity obtained from this model describes the experimental data[4, 9] well, whereas the previously proposed order parameters do not.

## I. NODAL SUPERCONDUCTIVITY IN $\text{PrOs}_4\text{Sb}_{12}$

Following Ref. [6], we consider energy gap functions  $\Delta(\mathbf{k})$  with point nodes at  $[0\ 1\ 0]$  and  $[0\ -1\ 0]$  in the B-phase, and with point nodes at  $[1\ 0\ 0]$ ,  $[0\ 1\ 0]$ ,  $[-1\ 0\ 0]$ , and  $[0\ -1\ 0]$  in the A-phase. Furthermore, in order to consistently describe the thermal conductivity data of Ref. [9] one also needs nodes at  $[0\ 0\ 1]$  and  $[0\ 0\ -1]$ . These constraints suggest

$$\vec{\Delta}_A(\mathbf{k}) = \frac{3}{2}\hat{d}\Delta e^{\pm i\phi_1 \pm i\phi_2 \pm i\phi_3} \left(1 - \hat{k}_1^4 - \hat{k}_2^4 - \hat{k}_3^4\right), \quad (1)$$

with  $e^{\pm i\phi_1} = \hat{k}_2 \pm i\hat{k}_3$  etc. for the A-phase, and with

$$\vec{\Delta}_B(\mathbf{k}) = \hat{d}\Delta e^{\pm i\phi} \left(1 - \hat{k}_3^4\right) \quad (2)$$

for the B-phase. We note that the p+h order parameter for the A phase satisfies the cubic symmetry. Also, for the B-phase the symmetry axis is rotated parallel to the crystal c-axis. Hence, the angular part,  $f = 1 - \hat{k}_3^4$ , of the B-phase is the same as for the previously considered s+g superconductor, whereas the angular part of the A-phase,  $f = \frac{3}{2} \left(1 - \hat{k}_1^4 - \hat{k}_2^4 - \hat{k}_3^4\right)$ , is different. The magnitude of these superconducting order parameters are shown in Fig. 1. Since both of these triplet order parameters break chiral symmetry, the ground state of the A-phase is sixfold degenerate and the ground state of the B-phase is twofold degenerate.

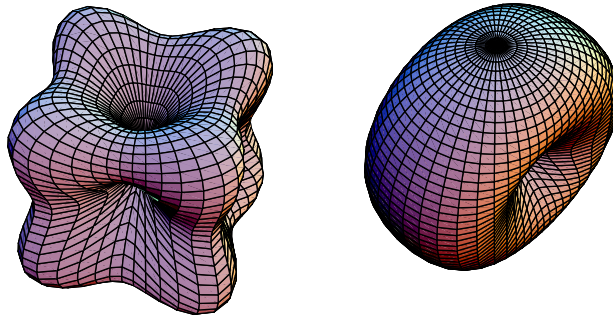


FIG. 1:  $|\Delta(\mathbf{k})|$  of the proposed p+h-wave superconducting order parameter in the A-phase (left) and in the B-phase (right) of  $\text{PrOs}_4\text{Sb}_{12}$ .

## II. QUASIPARTICLE SPECTRUM

In the absence of an external magnetic field, the quasiparticle density of states is given by

$$g(E) = |x| \text{Re} \left\langle \frac{1}{\sqrt{x^2 - |f|^2}} \right\rangle, \quad (3)$$

where  $x \equiv E/\Delta$ ,  $f = \frac{3}{2} (1 - \hat{k}_1^4 - \hat{k}_2^4 - \hat{k}_3^4)$  for the A-phase, and  $f = 1 - \hat{k}_3^4$  for the B-phase. Here  $\langle \dots \rangle = (4\pi)^{-1} \int d\Omega \dots$  denotes the angular average. These quasiparticle densities of states are evaluated numerically and shown in Fig. 2. In particular for the low-energy limit  $|x| \ll 1$  we find  $g(E) \approx \pi|x|/4$  for the A-phase and  $g(E) \approx \pi|x|/8$  for the B-phase.

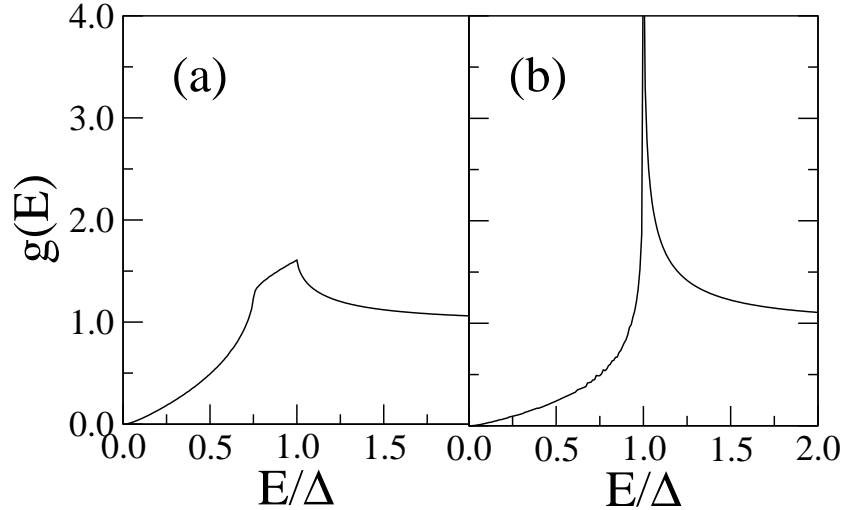


FIG. 2: Quasiparticle density of states in a p+h-wave superconductor. (a) A-phase, and (b) B-phase.

In the vortex state the quasiparticle density of states at  $E = 0$  is given by  $\pi \langle |\vec{v} \cdot \vec{q}| \rangle / (4\Delta)$  (A-phase) and  $\pi \langle |\vec{v} \cdot \vec{q}| \rangle / (8\Delta)$  (B-phase), where  $\vec{v} \cdot \vec{q}$  is the Doppler shift.[14, 15] Note that in this case  $\langle \dots \rangle$  denotes the average over the unit cell of the vortex lattice and over the nodal lines and points on the Fermi surface. For the field dependence we find

$$g_A(\vec{H}) = \frac{1}{2} \frac{v\sqrt{eH}}{\Delta} I_A(\theta, \phi) \quad (\text{A-phase}), \quad (4)$$

$$g_B(\vec{H}) = \frac{1}{4} \frac{v\sqrt{eH}}{\Delta} I_B(\theta, \phi) \quad (\text{B-phase}), \quad (5)$$

with

$$I_A(\theta, \phi) = \sin \theta + (1 - \cos^2 \theta \cos^2 \phi)^{1/2} + (1 - \cos^2 \theta \sin^2 \phi)^{1/2}, \quad (6)$$

$$I_B(\theta, \phi) = \sin \theta. \quad (7)$$

For the B-phase it was assumed here that the point nodes are aligned parallel to [001].

The corresponding specific heat, the spin susceptibility, and the superfluid density in the clean limit and at ultra-low temperatures are given by [16]

$$C_S/(\gamma_N T) = g(\vec{H}), \quad (8)$$

$$\chi_s/\chi_N = g(\vec{H}), \quad (9)$$

$$\rho_s(H)/\rho_S(0) = 1 - g(\vec{H}) \quad (\text{A - phase}), \quad (10)$$

$$\rho_s(H)/\rho_S(0) = 1 - 3g(\vec{H}) \quad (\text{B - phase}). \quad (11)$$

The superfluid density in the A-phase is isotropic and given by Eq. 10. However, in the B-phase Eq. 11 is only valid if the supercurrent flows parallel to the z axis (parallel to the nodal direction). Therefore it will be of great interest to study the Knight shift in the vortex state.

### III. ANGLE DEPENDENT MAGNETOTHERMAL CONDUCTIVITY

In order to analyze the thermal conductivity it is necessary to consider the effects of impurity scattering. Unlike for the s+g order parameter, the effects of disorder in the p+h-wave superconductor are more conventional.[17, 18] Here we consider impurity scattering in the unitary limit.

Let us first focus on the self-consistent equation for impurity scattering, given by  $iC_0\Delta = \lim_{\omega \rightarrow 0} \tilde{\omega}$ , where  $\tilde{\omega}$  is the disorder-renormalized frequency.[15] For the A-phase, this leads to

$$C_0 = \frac{2\Gamma}{\Delta} \left( C_0 \langle \ln \left( \frac{2}{\sqrt{C_0^2 + x^2}} \right) \rangle + \langle x \tan^{-1} \frac{x}{C_0} \rangle \right)^{-1}, \quad (12)$$

where  $\Gamma$  is the quasiparticle scattering rate in the normal state, and  $x \equiv |\vec{v} \cdot \vec{q}|/\Delta$ . For the B-phase the prefactor 2 on the right-hand side of the above equation is replaced by 4. In the superclean limit  $\langle x \rangle \gg C_0$  Eq. 12 leads to

$$C_{0A} = \frac{4\Gamma}{\pi\Delta} \langle x \rangle^{-1} \quad (\text{A - phase}), \quad (13)$$

$$C_{0B} = \frac{8\Gamma}{\pi\Delta} \langle x \rangle^{-1} \quad (\text{B - phase}) \quad (14)$$

On the other hand, in the clean limit we arrive at

$$C_{0A}^2 \ln \frac{2}{C_{0A}} = \frac{2\Gamma}{\Delta} - \frac{\langle x^2 \rangle}{2}, \quad (\text{A - phase}) \quad (15)$$

$$C_{0B}^2 \ln \frac{2}{C_{0B}} = \frac{4\Gamma}{\Delta} - \frac{\langle x^2 \rangle}{2}, \quad (\text{B - phase}). \quad (16)$$

In the absence of a magnetic field, the thermal conductivity exhibits universal heat conduction[19, 20] with  $\kappa_{00}/T = (\pi^2 n)/(12m\Delta)$  in the A-phase. In the B-phase there is universal heat conduction  $\kappa_{00}/T = (\pi^2 n)/(8m\Delta)$  only when the point nodes are aligned parallel to the heat current  $J_{\mathbf{q}}$ . Here  $\kappa_{00}$  is the thermal conductivity in the limits  $T \rightarrow 0$  and  $\Gamma \rightarrow 0$ . Hence the experimental data of Ref. [9] are consistent with p+h-wave superconductivity in  $\text{PrOs}_4\text{Sb}_{12}$ , but inconsistent with a s+g-wave order parameter.

In the vortex state, the thermal conductivity in the superclean limit is given by

$$\frac{\kappa_{zz}}{\kappa_N} = \frac{v^2(eH)}{8\Delta^2} \sin^2 \theta \quad (\text{A - phase}), \quad (17)$$

$$\frac{\kappa_{zz}}{\kappa_N} = \frac{3v^2(eH)}{64\Delta^2} \sin^2 \theta \quad (\text{B - phase}), \quad (18)$$

where  $J_{\mathbf{q}} \parallel z$ . Here the thermal conductivity in the normal state is given by  $\kappa_N = (\pi^2 nT)/(3m\Gamma)$ . For the B-phase we have assumed that the nodes are aligned to  $[0\ 0\ 1]$ . When the nodes are along  $[1\ 0\ 0]$  or  $[0\ 1\ 0]$ ,  $\kappa_{zz}$  is at least smaller by a factor of 10~50. Note that  $\kappa_{zz} \sim H \sin^2 \theta$  for both the A-phase and the B-phase. Therefore, the observed H-linear thermal conductivity  $\kappa_{zz}$  at  $T < 0.3K$  [9] follows from Eqs. 17 and 18. This implies that the crystals used in these measurements are in the superclean limit at sufficiently low temperatures.

In the clean limit the expressions for the thermal conductivity become

$$\frac{\kappa_{zz}}{\kappa_{00}} = 1 + \frac{3v^2(eH)}{40\Gamma\Delta} \ln \left( \sqrt{\frac{2\Delta}{\Gamma}} \right) \sin^2 \theta \ln \left( \frac{\Delta}{v\sqrt{eH} \sin \theta} \right) \quad (\text{A - phase}), \quad (19)$$

$$\frac{\kappa_{zz}}{\kappa_{00}} = 1 + \frac{v^2(eH)}{12\Gamma\Delta} \ln \left( \sqrt{\frac{2\Delta}{\Gamma}} \right) \sin^2 \theta \ln \left( \frac{\Delta}{v\sqrt{eH} \sin \theta} \right) \quad (\text{B - phase}), \quad (20)$$

where  $\kappa_{00}$  is the thermal conductivity in the limit of universal heat conduction. Hence, in the clean limit the field-dependent part of  $\kappa_{zz}$  is given by  $\kappa_{zz} \sim HF(\theta)$ , where  $F(\theta) = \sin^2 \theta \ln(C/\sin \theta)$  and  $C = \Delta/v\sqrt{eH}$ . From these equations we observe that the  $\theta$ -dependence of the leading terms is the same for the A-phase and B-phase. In Fig. 3, we plot the measured  $\theta$ -dependence of  $\kappa_{zz}$  in  $\text{PrOs}_4\text{Sb}_{12}$  [9] at various applied magnetic fields

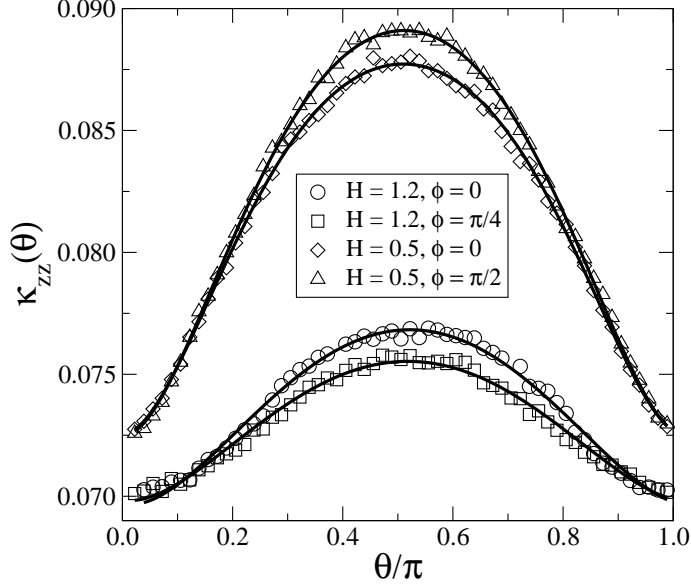


FIG. 3: Angle dependence of the thermal conductivity in an applied magnetic field. The symbols are experimental data from Ref. [9], and the solid lines are fits to  $HF(\theta)$ , as discussed in the text.

and azimuthal angles  $\phi$ . Fits of this data to  $HF(\theta)$  are shown as solid lines, suggesting that these samples are in the clean limit. In these fits  $C = 5$  and  $C = 3$  are obtained for  $H = 0.5T$  and  $H = 1.2T$  respectively. Thus, using the weak-coupling theory gaps  $\Delta_A = 4.2K$  and  $\Delta_B = 3.5K$  for the A- and B-phase, we deduce  $v = 0.96 \times 10^7 cm/sec$  and  $\Gamma \simeq 0.1K$ . These values are reasonable,[22] indicating that the quasiclassical approximation is reliable.

#### IV. CONCLUDING REMARKS

In this work, we have proposed a spin-triplet p+h-wave order parameter to account for observed features in the superconducting phases of  $PrOs_4Sb_{12}$ . This model describes well the angle dependent thermal conductivity data by Izawa et al., Refs. [4] and [9]. In order to be fully consistent, we have discovered that the nodal directions of  $\Delta(k)$  in the B-phase have to be aligned parallel to the external magnetic field in the field-cooled configuration. This triplet superconductivity in  $PrOs_4Sb_{12}$  is not surprising since many other heavy-fermion superconductors appear to have spin-triplet order parameters, including  $UPt_3$ ,  $UBe_{13}$ ,  $URu_2Si_2$ , and  $UNi_2Al_3$ . [21] The interesting dependence of the nodal points in  $\Delta(k)$  on the external magnetic field deserves further study.

We are grateful to H. Tou for useful correspondence about his NMR measurements on

PrOs<sub>4</sub>Sb<sub>12</sub>, and to P. Thalmeier for useful conversations. K.M. is grateful for the hospitality of the Max-Planck Institute for the Physics of Complex Systems at Dresden, where part of this work was performed. S.H. acknowledges financial support by the NSF under Grant No. DMR-0089882.

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  - [9] K. Izawa et al., unpublished. This study contains two sets of measurements of  $\kappa_{zz}$ . The first set is only for  $H \parallel c$  and  $H \parallel b$ , but taken down to 150mK. Both the T and H linear dependences for  $T < 0.3\text{K}$  are consistent with the superclean limit formulas (Eqs. 17 and 18). The second set of data is taken at 0.3K in a magnetic field rotated within the x-z plane. The observed theta dependence of  $\kappa_{zz}$  are consistent with the clean limit formulas (Eqs. 19 and 20).
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- [22] From de Haas-van Alphen measurements (H. Sugawara, S. Osaki, S. R. Saha, Y. Aoki, H. Sato, Y. Inada, H. Shishido, R. Settai, Y. Onuki, H. Harima, and K. Oikawa, Phys. Rev. B **66**, 220504(R) (2002))  $v$  is estimated to be  $0.7 \times 10^7 cm/sec$  ( $\alpha$ -band),  $0.66 \times 10^7 cm/sec$  ( $\beta$ -band), and  $0.23 \times 10^7 cm/sec$  ( $\gamma$ -band).